Strong Electronic Correlations in Aqueous Actinide Complexes

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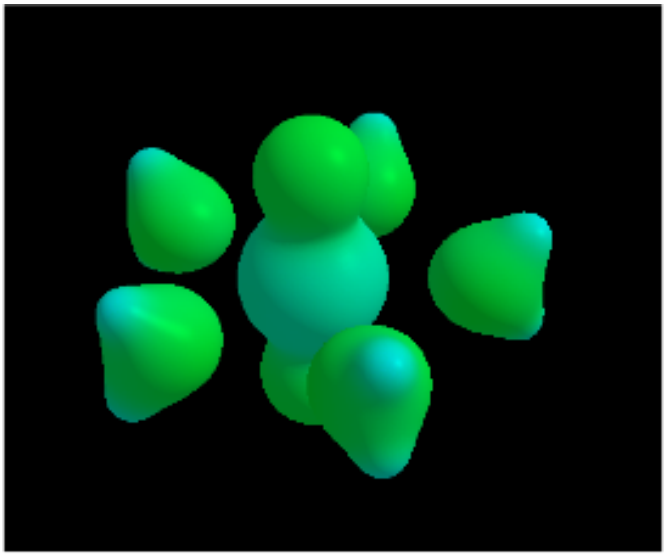
We are working on understanding the quantum chemistry of aqueous actinide complexes. Strong electronic correlations likely play an important role, due to the relatively localized nature of the actinide 5f orbitals. As a first step density-functional calculations are being carried out that incorporate relativity as well as solvation effects. Reduced Hubbard type models will then be contructed to incorporate the large Coulomb repulsion; the models will be solved by exact diagonalization and by mean-field methods. The conjunction of nanoscale complexification, an aqueous environment, and strong electronic correlations makes this a particularly fascinating and challenging problem.



Relativistic density-functional calculation of a uranium dioxide molecule surrounded by 5 water molecules. Bond lengths so obtained agree well with existing Extended X-ray Absorption Fine Structure (EXAFS) measurements.

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Broader Impacts: Our work will be applied to the problem of environmental dispersal of plutonium and neptunium from nuclear waste storage sites such as the one proposed for Yucca Mountain, Nevada. Disproportionation into multiple oxidation states, with widely varying solubilities, complicates geochemical transport mechanisms and is poorly understood at present.

Education: Three graduate students (Steve Horowitz, Sootaek Lee, and Ookie Ma) are supported in part by the grant. Horowitz is carrying out the DFT calculations and will also work on the construction and solution of the effective Hubbard models.

Outreach: The PI has been giving a talk called "The Quantum Mechanics of Global Warming" both as a physics colloquium and as a public lecture (7 times during the past year).